

Interference of Mesoscopic Particles: Quantum–Classical Transition

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Abstract. We analyze the double slit interference of a mesoscopic particle. We calculate the visibility of the interference pattern in the far field case, introduce a characteristic temperature that defines the onset to decoherence and scrutinize the conditions that must be satisfied for an interference experiment to be possible.

1. Introduction

Interference is one of the most characteristic traits of quantum systems. As Dirac clarified [1], this phenomenon is rooted in the superposition principle, according to which different states of a single quantum mechanical particle interfere with each other. The simplest case is that of two states: double slit interference has been observed with photons, electrons, neutrons, atoms and small molecules [2], and recently even with large molecular clusters [3]. Our comprehension of the quantum mechanical world has been shaped, to a large extent, by the ideas that motivated these experiments.

Quantum particles interfere, but classical particles do not, and it is not easy to understand where the borderline has to be placed. The size of the interfering system plays an important role, but it is certainly not the only relevant variable: for example, double slit interference has been observed with molecules, but not

with protons. In this article we will analyze the interference of mesoscopic systems, endowed with an internal structure which leads to entanglement with their environment (e.g., via photon emission). An example is a fullerene molecule flying between a diffraction grating and a detector. Some classical features are apparent for such mesoscopic systems, yet their ability to interfere is preserved, at least to some extent, and can be viewed as a quantum signature. The main objective of this article will be to understand under which conditions a “large” system interferes and which of its dynamical variables can interfere.

2. Double Slit Interference in a Poissonian Environment

Let us consider a mesoscopic quantum system (molecule), whose center of mass is described by a (double) wave packet $|\psi_0\rangle$, emerging from a double slit. The wave packet travels along direction $+z$; the slits are parallel to y and are separated by a distance d along direction x

$$|\psi_0\rangle = \frac{1}{\sqrt{2}}(|\psi_+\rangle + |\psi_-\rangle), \quad |\psi_\pm\rangle = \exp(\pm idp_x/2)|\psi_{\text{slit}}\rangle, \quad (1)$$

p_x being the x component of the momentum operator and $|\psi_{\text{slit}}\rangle$ the state emerging from one slit ($\hbar = 1$). We assume that $\langle\psi_-|\psi_+\rangle = 0$, so that $|\psi_0\rangle$ is normalized. During its travel to the screen, the molecule emits photons and recoils accordingly. The internal state of the molecule together with the photon field plays therefore the role of environment: such an environment disturbs the motion of the center of mass, via scattering processes (typically photon emissions, yielding momentum kicks). We shall assume that the internal temperature of the mesoscopic system is much higher than the temperature of the photon field. This hypothesis, verified e.g. in present day experiments with fullerenes, will be discussed in the next section.

Let the molecule undergo momentum kicks due to photon emissions. The Hamiltonian describing the evolution of the x component of the center of mass in the presence of random kicks Δp_k at times t_k reads (henceforth, for simplicity, $p_x = p$)

$$H_\xi(t) = \frac{p^2}{2m} - \xi(t)x, \quad \xi(t) = \sum_k \delta(t - t_k)\Delta p_k, \quad (2)$$

where $\{t_k\}_{k \in \mathbb{Z}}$ is a shot noise with density Λ and the momentum jumps Δp_k are identically distributed independent random variables with probability density $W(\Delta p_k)$. The process $\xi(t)$ is the time derivative of a compound Poisson process [4]. Both Λ and W are functions of the state of the environment (for example of its temperature T). The time evolution of a wave packet that emerges at time $t_0 = 0$ from the slits reads

$$|\psi(t)\rangle = U_\xi(t)|\psi_0\rangle, \quad (3)$$

$U_\xi(t)$ being the unitary evolution engendered by the Hamiltonian (2)

$$\begin{aligned}
 U_\xi(t) &= \mathcal{T} \exp \left(-i \int_0^t ds H_\xi(s) \right) \\
 &= e^{-i(t-t_n)p^2/2m} \mathcal{T} \prod_{k=1}^n e^{ix\Delta p_k} e^{-i\Delta t_{k-1}p^2/2m} \\
 &= e^{ix\Delta p^{(n)}} \prod_{k=0}^n e^{-i(p-\Delta p^{(k)})^2 \Delta t_k/2m}, \tag{4}
 \end{aligned}$$

where $\Delta t_k = t_{k+1} - t_k$, $\Delta p^{(k)} = \sum_{j=1}^k \Delta p_j$ is the total effect of k momentum jumps ($\Delta p^{(0)} = 0$), the total number of collisions n is a Poisson random variable with mean Λt , and \mathcal{T} is the time-ordering operator, forcing earlier times (lower k) at the right. In the third equality we used the commutation relation

$$e^{-ip^2 \Delta t/2m} e^{ix\Delta p} = e^{ix\Delta p} e^{-i(p-\Delta p)^2 \Delta t/2m},$$

in order to move all kick operators to the far left side. From (3) and (4) one gets

$$\begin{aligned}
 \psi(x, t) &= \langle x | \psi(t) \rangle = e^{ix\Delta p^{(n)}} \langle x | e^{-i \sum (p-\Delta p^{(k)})^2 \Delta t_k/2m} | \psi_0 \rangle \\
 &= e^{ix\Delta p^{(n)}} \int \frac{dp}{\sqrt{2\pi}} e^{-i\phi(p)} \tilde{\psi}_0(p), \tag{5}
 \end{aligned}$$

where $\tilde{\psi}_0(p) = \langle p | \psi_0 \rangle$ and

$$\phi(p) = \sum_{k=0}^n \frac{(p - \Delta p^{(k)})^2 \Delta t_k}{2m} - xp = \phi(\bar{p}) + \frac{t}{2m} (p - \bar{p})^2, \tag{6}$$

with $t = \sum \Delta t_k$ and \bar{p} the value of the momentum at the extremal $\phi'(\bar{p}) = 0$,

$$\bar{p}(x, t) = \frac{mx}{t} + \sum_{k=0}^n \frac{\Delta p^{(k)} \Delta t_k}{t} = \frac{mx}{t} + \sum_{k=1}^n \zeta_k \Delta p_k, \tag{7}$$

where $\zeta_k = 1 - t_k/t$ characterize the emissions between the slits and the screen. A peculiarity of this analysis is the presence of the *same* Poisson process on *both* branch waves: this is due to our assumption that the most relevant part of the environment (as far as we focus on decoherence) is the internal state of the mesoscopic molecule, emitting (thermal) long-wavelength photons into the background field; by contrast, for an “external” environment (such as a high pressure gas in which the molecule travels), one should have considered *two independent* Poisson processes, one for each branch wave.

Equation (5) represents the convolution of the initial momentum wave packet with a Gaussian

$$\psi(x, t) = e^{i[x\Delta p^{(n)} - \phi(\bar{p})]} \int \frac{dp}{\sqrt{2\pi}} e^{-i\frac{t}{2m}(p-\bar{p})^2} \tilde{\psi}_0(p), \tag{8}$$

whose spread $(m/t)^{1/2}$ becomes narrower as time t increases. For $t \rightarrow \infty$ (8) reads

$$\psi(x, t) \sim e^{i[x\Delta p^{(n)} - \phi(\bar{p})]} \left(\frac{m}{it}\right)^{\frac{1}{2}} \tilde{\psi}_0(\bar{p}). \quad (9)$$

This approximation is valid for $t \gg m|\tilde{\psi}_0''(\bar{p})/\tilde{\psi}_0(\bar{p})|$ and implies that $\tilde{\psi}_0(p)$ be evaluated at $p = \bar{p}(x, t)$, at the screen. In the following we will always suppose that such condition holds (far-field interference pattern). The interference pattern reads

$$I(x, t) = \langle |\psi(x, t)|^2 \rangle \sim \left(\frac{m}{t}\right) \langle |\tilde{\psi}_0(\bar{p})|^2 \rangle, \quad (10)$$

where $\langle \dots \rangle$ denotes the average over the process $\xi(t)$. With the initial state (1), $\tilde{\psi}_0(p) = \sqrt{2}\tilde{\psi}_{\text{slit}}(p) \cos(pd/2)$, with $\tilde{\psi}_{\text{slit}}(p) \equiv \langle p|\psi_{\text{slit}}\rangle$, and the far-field condition is satisfied for $t \gg md^2$. Under this condition, the intensity at the screen reads

$$I(x, t) = \left(\frac{m}{t}\right) \left| \tilde{\psi}_{\text{slit}}\left(\frac{mx}{t}\right) \right|^2 \left[1 + \langle \cos(\bar{p}d) \rangle \right], \quad (11)$$

where we approximated $|\tilde{\psi}_{\text{slit}}(\bar{p})|^2 \simeq |\tilde{\psi}_{\text{slit}}(mx/t)|^2$, for weak enough kicks. The corresponding visibility \mathcal{V} is defined by

$$\begin{aligned} I(x, t) &= I_0(x, t) \left[1 + \mathcal{V} \cos\left(\frac{md}{\hbar t}x + \phi\right) \right], \\ \mathcal{V} &= |F|, \quad \phi = \arg F, \\ F &= \left\langle \exp\left(id \sum_{k=1}^n \zeta_k \Delta p_k\right) \right\rangle = \mathcal{V} e^{i\phi}, \end{aligned} \quad (12)$$

where $I_0(x, t) = (m/t)|\tilde{\psi}_{\text{slit}}(mx/t)|^2$. In order to calculate the visibility, the features of the average $\langle \dots \rangle$ must be expressed in terms of the distribution of the momentum jumps $W(\Delta p_k)$ and the Poisson times $\{t_k\}$.

By performing first the average over $W(\Delta p_k)$ we get

$$F = \left\langle \prod_{k=1}^n g(t_k) \right\rangle_{\Lambda}, \quad (13)$$

where $g(t_k) = f(dt - t_k/t)$ and

$$f(x) = \langle \exp(ix\Delta p) \rangle_{\Delta p} = \int du W(u) \exp(ixu), \quad (14)$$

$\langle \dots \rangle_{\Lambda}$ denoting the average over the shot noise with density Λ . This is easily computed [5]

$$F = 1 + \sum_{n=1}^{\infty} \frac{1}{n!} \left[\Lambda \int_0^t (g(\tau) - 1) d\tau \right]^n = \exp\left(-\Lambda t \int_0^1 [1 - f(sd)] ds\right) \quad (15)$$

and yields the visibility

$$\mathcal{V} = \exp(-\Lambda\zeta t), \quad \zeta = \int_0^1 [1 - \operatorname{Re} f(sd)] ds, \quad (16)$$

where Re denotes the real part. By using the definition (14), the “geometrical” factor reads $\zeta = \langle 1 - \operatorname{sinc}(d\Delta p) \rangle_{\Delta p}$, where $\operatorname{sinc}(x) = \sin x/x$, and the visibility (16) can be given the useful expression

$$\mathcal{V} = \exp\left(-\Lambda t \langle 1 - \operatorname{sinc}(d\Delta p) \rangle_{\Delta p}\right). \quad (17)$$

Notice that $\mathcal{V} \leq 1$, because $\operatorname{sinc}(x) \leq 1$. Moreover, if the jumps are symmetrically distributed, i.e. $W(\Delta p) = W(-\Delta p)$, then $f(x)$ is a real function and one can omit the real part in (16), so that $\mathcal{V} = F$.

3. Thermodynamics

In order to calculate the visibility from (17) we need to evaluate the kick rate Λ and the probability density of the momentum jumps $W(\Delta p)$ for a large molecule at a given temperature. Planck’s blackbody formula is not valid for small atomic clusters and needs to be generalized on two counts. One is the finiteness of the number of modes (freedoms), the other is the reduced stimulated emission. The former influences the high-energy part of the spectrum, the latter the low-energy part. In addition, there are finite-size effects that need to be taken into account.

If the interfering cluster can be considered (almost) isolated during its travel to the screen, its temperature is in general not in equilibrium with that of the radiation field. When photon absorption from the background radiation can be neglected, e.g. because the temperature of the molecule is much higher than that of the background field (which is the case in which we are interested), the photon emission rate reads [6]

$$R_T(\omega) = \frac{\omega^2 \sigma_{\text{abs}}(\omega)}{\pi^2 c^2} \exp\left[-\frac{\hbar\omega}{k_B T} - \frac{k_B}{2C_V} \left(\frac{\hbar\omega}{k_B T}\right)^2\right], \quad (18)$$

where ω is the photon frequency, σ_{abs} and $C_V = Nk_B$ are the absorption cross section and heat capacity of the small particle, respectively, T its temperature and N the number of vibrational modes (for example, $N \simeq 170$ for C_{60} , $N \simeq 200$ for C_{70}).

The very notion of temperature for small atomic clusters is interesting and deserves a few words of explanation [6]. These mesoscopic particles exhibit a strong statistical mixing of states, so that their radiation emission features are independent of the excitation mechanism. This is a consequence of the rapidity that characterizes the energy exchange mechanisms between quasistationary states, compared to the slower radiative lifetime. The resulting effect is the generalization (18) of Planck’s formula, which accounts for the finite size of the cluster,

the limited heat capacity and the emission of radiation even without any external photon bath surrounding the system. These ingredients yield the sensible and consistent notion of molecular temperature given in (18).

The total photon emission rate reads

$$\Lambda(T) = \int_0^{\infty} d\omega R_T(\omega), \quad (19)$$

and we shall assume that the temperature of the molecule does not change appreciably due to photon emission during the flight. We will check the validity of this assumption later. In order to compute these quantities, we have to determine the ω dependence of the absorption cross section. We shall heuristically assume the simple form

$$\sigma_{\text{abs}}(\omega) = a_\ell \omega^\ell, \quad (20)$$

where ℓ is a positive integer and a_ℓ a real number, and look for the best fit. For instance, in the case of the fullerenes C_{60} and C_{70} , a comparison with experiment [7] yields accurate fits for $\ell = 4$, and $a_4 = 7.04 \times 10^{-66} \text{nm}^2 \text{s}^4$ and $a_4 = 7.79 \times 10^{-66} \text{nm}^2 \text{s}^4$, respectively. In general, care must be taken in order to reproduce the features of the absorption cross section in the visible and infrared region, rather than, say, in the ultraviolet one. This is because for the present generation of experiments [3] decoherence is mainly due to close-infrared photon emission. Clearly, the values of ℓ and a_ℓ will be different for other molecules, but our general conclusions will not change.

By plugging (20) into (18) one gets the series

$$R_T(\omega) = \frac{a_\ell \omega^{\ell+2}}{\pi^2 c^2} e^{-\frac{\hbar\omega}{k_B T}} \sum_{m=0}^{\infty} \frac{(-1)^m}{(2N)^m m!} \left(\frac{\hbar\omega}{k_B T} \right)^{2m} \quad (21)$$

and integrating term by term in (19) one gets the asymptotic expansion for large N

$$\Lambda(T) \sim \frac{a_\ell}{\pi^2 c^2} \left(\frac{k_B T}{\hbar} \right)^{\ell+3} \sum_{m=0}^{\infty} \frac{(2m + \ell + 2)!}{(-2N)^m m!}. \quad (22)$$

For fullerenes, typical emission rates at $T = 2500$ for a time of flight $t \simeq 2$ ms yields 4-5 emitted infrared photons during the flight in the interferometer. In such case, the temperature of the molecule decreases by just a few percent, which does not affect the emission rate, and corroborates our initial assumption (after (19)).

The momentum kick on the molecule after the emission of a photon of frequency ω has magnitude $|\mathbf{p}| = \hbar\omega/c$. By assuming that the emission process is isotropic, the probability density that the molecule undergoes a momentum jump \mathbf{p} reads

$$W^{(3D)}(\mathbf{p}) = \frac{1}{4\pi \mathbf{p}^2} \frac{c}{\hbar \Lambda(T)} R_T \left(\frac{c|\mathbf{p}|}{\hbar} \right) \quad (23)$$

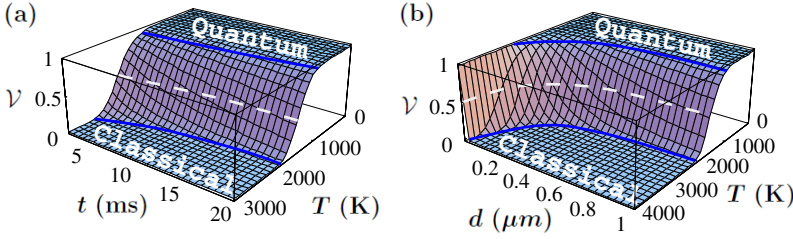


Fig. 1: (a): Visibility \mathcal{V} vs T (0–3000 K) and t (3–20 ms) for $d = 1\mu\text{m}$. (b): Visibility \mathcal{V} vs T (0–4000 K) and d (0.01–1 μm) for $t = 10$ ms. The white dashed line indicates the level curve $\mathcal{V} = 1/2$. We plot only the physically relevant ranges of the parameters.

and yields the one-dimensional probability density

$$W(\Delta p) = \int d^3p W^{(3D)}(\mathbf{p})\delta(p_x - \Delta p) = \frac{c}{2\hbar\Lambda(T)} \int_{c|\Delta p|/\hbar}^{\infty} \frac{d\omega}{\omega} R_T(\omega). \quad (24)$$

By plugging (24) into (14) we get

$$f(x) = \frac{1}{\Lambda(T)} \int_0^{\infty} d\omega R_T(\omega) \operatorname{sinc}\left(\frac{\omega x}{c}\right), \quad (25)$$

and from (16)

$$\mathcal{V}(T, d, t) = \exp(-[\Lambda(T) - G(T, d)]t), \quad (26)$$

where

$$G(T, d) = \int_0^1 ds \int_0^{\infty} d\omega R_T(\omega) \operatorname{sinc}\left(\frac{\omega d}{c}s\right). \quad (27)$$

Note that $G(T, d) \sim \Lambda(T)$, for $T \rightarrow 0$, so that for low temperatures $\mathcal{V} \rightarrow 1$. In the high temperature case, on the contrary, $\mathcal{V} \rightarrow 0$, as expected for a classical particle. By plugging the series (21) into (27) and integrating term by term one gets

$$G(T, d) \sim \frac{a_\ell}{\pi^2 cd} \left(\frac{k_B T}{\hbar}\right)^{\ell+2} \sum_{m=0}^{\infty} \left\{ \frac{(-1)^m (2m + \ell + 1)!}{(2N)^m m!} \times \int_0^{\frac{dk_B T}{\hbar c}} dx \frac{\sin\left[(2m + \ell + 2) \arctan x\right]}{x(1+x^2)^{(2m+\ell+2)/2}} \right\}. \quad (28)$$

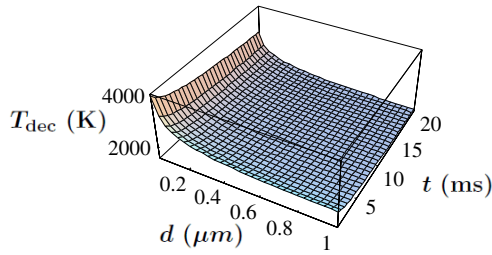


Fig. 2: T_{dec} vs. d (0.02–1 μm) and t (1–20 ms). We plot only the physically relevant ranges of the parameters.

4. Visibility and Quantum-Classical Transition

For the sake of concreteness, we shall focus on the fullerene molecule C_{70} , for which $N \simeq 200$, and $\ell = 4$, $a_4 = 7.79 \times 10^{-66} \text{nm}^2 \text{s}^4$, as explained after Eq. (20). By inserting the expansions (22) and (28) into the visibility (26) one gets the plots in Fig. 1(a) for a fixed distance between the slits, and in Fig. 1(b) for a fixed time of flight. A quantum system, characterized by the value $\mathcal{V} = 1$, tends to display a classical behaviour, characterized by $\mathcal{V} = 0$, when the time of flight and/or the distance between the slits are increased. This quantum-classical transition takes place at a “decoherence temperature” $T = T_{\text{dec}}$ determined by the level curve (white dashed line in Fig. 1)

$$\mathcal{V} = 1/2 \iff [\Lambda(T_{\text{dec}}) - G(T_{\text{dec}}, d)]t - \ln 2 = 0. \quad (29)$$

$T_{\text{dec}}(d, t)$ is plotted in Fig. 2. The transition between the quantum and classical behaviour is very sharp, both in Figs. 1(a) and 1(b), and this enables us to define the decoherence temperature in a clear-cut way. These graphs are the central results of our analysis.

Our conclusions are valid under two implicit assumptions: i) scattering between the mesoscopic molecule and the molecules of the surrounding gas can be neglected, and ii) the temperature of the molecular beam can be considered uniform (i.e., it does not vary much among the molecules detected in one experimental run). The first hypothesis is easily seen to be very accurate, at least in air at ordinary pressures, and will not be discussed here (see [8] for an accurate analysis of the near-field case with different gases). The second hypothesis is more delicate, as in some experiments the mesoscopic particle excitation/heating process by a multiple laser beam system can be position- and/or velocity-dependent (e.g., see the second experiment in [3]), yielding temperature distributions with variations of tens of percent (see [9] for an analysis of the near-field case). In such a case one should take an additional temperature average in (26). This average is heavily experiment dependent, and would require a careful estimate of the heating process. Such a problem will be tackled in the future. At any rate, it does not modify the conclusions of the present article. Finally, notice that H in (2) is the semiclassical approximation of a fully quantum mechanical (QED) Hamiltonian, in which pho-

ton emission processes (and consequent recoils) are explicitly taken into account [10]. In this sense, our analysis neglects entanglement effects and yields therefore conservative estimates of the decoherence process.

5. Remarks on the Interference of a Mesoscopic Particle

A mesoscopic system, such as a macromolecule, can be attributed a temperature, in the sense of (18), by virtue of its large number of freedoms N . In a double slit interference experiment, the degree of freedom associated with the interfering pattern [the relevant variable being x , see (1)] plays a special role. We now argue that, in general, such an “interfering” freedom is not in the same thermal state as the others.

Let the experiment last for a time t (the time of flight of the molecule in the interferometer) and H_{exch} be the interaction Hamiltonian responsible for the coupling between the interfering freedom and the environment. Interference can be observed if

$$\left\langle \int_0^t H_{\text{exch}} dt \right\rangle_T \lesssim \hbar \ll k_B T t, \quad (30)$$

where the average $\langle \dots \rangle_T$ is taken over the initial state of the total system. The above one is a condition on the exchanged action and the environmental temperature. For example, if the average in (30) is understood in the r.m.s. sense and $H_{\text{exch}} = -\xi(t)x$, like in (2), we have $\langle \int H_{\text{exch}} dt \rangle = \sqrt{\Lambda t} \langle \Delta p \rangle d = \Delta p_{\text{tot}} d \leq \hbar$ where $\Delta p_{\text{tot}}(T)$ is the total recoil due to a momentum random walk [11]. In such a case the first inequality in (30) is nothing but (the opposite of) Rayleigh’s criterion in Heisenberg’s microscope and this clarifies the rationale behind it.

It is worth stressing again that the temperature of the photon bath has been neglected, see comments after (1): strictly speaking our environment is made up of two parts (the other freedoms of the molecule + the photon bath), that are not in equilibrium, and T is the “local” temperature of the roto-vibrational and electronic modes of the macromolecule, responsible for photon emission. We are not considering the (much) slower equilibration process of the whole environment.

When (30) is satisfied, the macromolecule interferes. During the interference experiment, energy flows between the environment and the interfering “colder” freedom. Such a freedom, associated with the interfering component p_x , approaches equilibrium (at a temperature T), via momentum — and energy — transfer, during the momentum random walk process described above: eventually, the visibility vanishes and interference is lost when the first inequality in (30) ceases to be valid. This is a rather fast process, that induces a classical behaviour in the (relevant interfering variable of the) mesoscopic system. The thermalization process sets in afterwards, when $\langle \int H_{\text{exch}} dt \rangle \simeq k_B T t \gg \hbar$ instead of (30), and is much slower. This is, altogether, a remarkable picture, that adds spell to the interfering features of these mesoscopic systems, as well to the many additional

problems that must be considered [12] in order to get a complete picture of these phenomena.

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